

S.-C. Chou, et al.
USSN: 10/047,835
Page 2

Amendments To The Claims:

This listing of claims will replace all prior versions, and listings, of claims in the application:

Listing of claims:

1. (presently amended) A method for recovering and producing C₄-C₆ dicarboxylates from an alkaline waste solution generated in a caprolactam preparation process, comprising the steps of:

- (1) neutralizing adding sulfuric acid to the alkaline waste solution generated from the caprolactam preparation process and to adjusting a pH value thereof with sulfuric acid to a pH of less than 7, to separatinge the alkaline waste solution into an aqueous phase and an organic phase; adding nitric acid to the organic phase for oxidizing and converting valuable substances selected from adipic acid, 6-hydroxycaproic acid, saponified esters or ketones of cyclohexanol, and C₄-C₆ lactons, contained in the organic phase into dicarboxylic acids, thereby obtaining an oxidized reaction mixture containing dicarboxylic acids;
- (2) introducing the oxidized reaction mixture obtained from the step (1) into a two-stage concentration apparatus; in first-stage concentration, distilling out low boiling-point monocarboxylic acids and nitric acid; in second-stage concentration, decomposing remaining nitric acid and nitrocompounds, so as to obtain crude concentrates mainly containing C₄-C₆ dicarboxylic acids;
- (3) adding C₁-C₄ alkyl alcohol to the crude concentrates mainly containing C₄-C₆ dicarboxylic acids obtained from the step (2), and proceeding two-stage esterification, whereby half-esterified intermediates are obtained in first-stage esterification, and crude dicarboxylates are obtained in second-stage esterification; and

S.-C. Chou, et al.
USSN: 10/047,835
Page 3

(4) distilling the crude dicarboxylates obtained from the step (3), so as to get single-species dicarboxylates or a mixture of dicarboxylates.

2. (presently amended) The method of claim 1, wherein in the step (1), at least one oxidant other than nitric acid selected from the group consisting of hydrogen peroxide, perchloric acid, and potassium permanganate, is added to the organic phase.

3. (canceled).

4. (original) The method of claim 1, wherein concentration of nitric acid is 10 to 90%.

5. (original) The method of claim 1, wherein a weight ratio of nitric acid to the organic phase is 0.5 to 30:1.

6. (original) The method of claim 2, wherein the oxidant is added in a ratio of 0 to 5%.

7. (original) The method of claim 1, wherein oxidation and conversion of the step (1) are performed under conditions including: at least one reaction stage, reaction temperature of a range from 10 to 150°C, and reaction mixture flowing from first to last reaction stage with the reaction temperature being increased gradually at stage intervals of 5 to 30°C, reaction time for each stage being set as 5 minutes to 4 hours, and reaction pressure being absolute pressure 0.5 to 2kg/cm².

8. (presently amended) The method of claim 1, wherein in the step (2), the first-stage concentration is performed at a temperature of 50 to 120°C and an absolute pressure of 0.2

S.-C. Chou, et al.
USSN: 10/047,835
Page 4

to 1.5 kg/cm², the second-stage concentration is carried out at a temperature of 120 to 200°C and an absolute pressure of 0.5 to 2.0 kg/cm².

9. (original) The method of claim 1, wherein the C₁-C₄ alkyl alcohol used in the step (3) is selected from a group consisting of methanol, ethanol, propanol and butanol.

10. (original) The method of claim 1, wherein a weight ratio of the C₁-C₄ alkyl alcohol to the crude concentrates of dicarboxylic acids is 1-15:1.

11. (original) The method of claim 9, wherein a weight ratio of the C₁-C₄ alkyl alcohol to the crude concentrates of dicarboxylic acids is 1-15:1.

12. (original) The method of claim 1, wherein a catalyst is added in the step (3), and selected from a group consisting of sulfuric acid, phosphoric acid, nitric acid, alkanesulfonic acids, benzenesulfonic acid and cation exchange resin.

13. (original) The method of claim 12, wherein the catalyst is added in an amount of 0 to 5%.

14. (presently amended) The method of claim 1, wherein in the step (3), the first-stage esterification is performed at a temperature of 40 to 120°C, an absolute pressure of 0.2 to 1.2 kg/cm², and esterification time of 0.5 to 8 hours; and the second-stage esterification is carried out at a temperature of 80 to 200°C, an absolute pressure of 0.8 to 2.5 kg/cm², and esterification time of 0.5 to 8 hours.

15. (original) The method of claim 1, wherein a distillation apparatus used in the step (4) is of tray or packing type.

S.-C. Chou, et al.
USSN: 10/047,835
Page 5

16. (original) The method of claim 14, wherein distillation of the step (4) is performed by using 10 to 100 trays theoretically, and under absolute pressure of 0.02 to 1.0 kg/cm² and temperature of 70 to 250°C.

17. (presently amended) The method of claim 1, comprising the steps of:

- (1) adding sulfuric acid to neutralizing the alkaline waste solution generated from the caprolactam preparation process to adjust and adjusting a pH value thereof to a pH of less than 7, and separating the alkaline waste solution with sulfuric acid to separate into an aqueous phase and an organic phase; adding nitric acid of 10 to 90% concentration to the organic phase in a weight ratio of nitric acid to the organic phase at 0.5 to 30:1, and oxidizing and converting valuable substances selected from adipic acid, 6-hydroxycaproic acid, saponified esters or ketones of cyclohexanol, and C₄-C₆-lactones contained in the organic phase into dicarboxylic acids, thereby obtaining an oxidized reaction mixture containing dicarboxylic acids;
- (2) introducing the oxidized reaction mixture obtained from the step (1) into a two-stage concentration apparatus, wherein first-stage concentration is performed at a temperature of 50 to 120°C and an absolute pressure of 0.2 to 1.5 kg/cm², and second-stage concentration is carried out at a temperature of 120 to 200°C and an absolute pressure of 0.5 to 2.0 kg/cm², so as to obtain crude concentrates mainly containing C₄-C₆ dicarboxylic acids;
- (3) adding C₁-C₄ alkyl alcohol to the crude concentrates mainly containing C₄-C₆ dicarboxylic acids obtained from the step (2), and proceeding two-stage esterification, wherein first-stage esterification is performed at temperature of 40 to 120°C, absolute pressure of 0.2 to 1.2 kg/cm², and esterification time of 0.5 to 8 hours; and second-stage esterification is carried out at temperature of 80 to 200°C, absolute pressure of 0.8 to 2.5 kg/cm², and esterification time of 0.5 to 8 hours, so as to obtain crude dicarboxylates; and

S.-C. Chou, et al.
USSN: 10/047,835
Page 6

- (4) introducing the crude dicarboxylates obtained from the step (3) into a tray- or packing-type distillation tower for performing distillation by using 10 to 100 trays theoretically, and under absolute pressure of 0.02 to 1.0 kg/cm² and temperature of 70 to 250°C, so as to get single-species dicarboxylates or a mixture of dicarboxylates.